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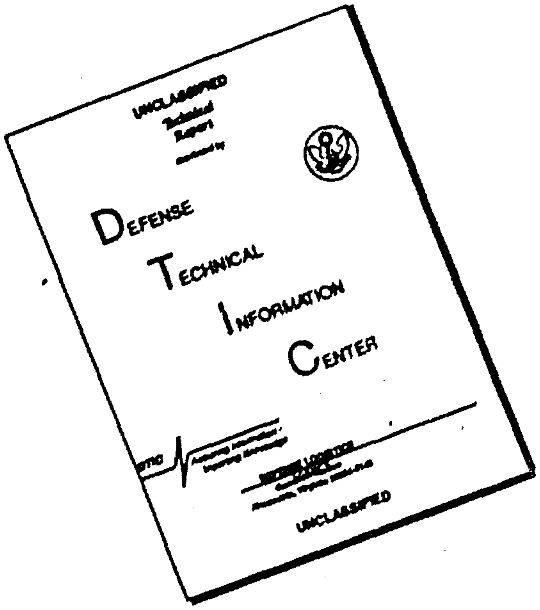

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USNRDL-TR-680
12 September 1963

FRACTIONATION III.

ESTIMATION OF DEGREE OF FRACTIONATION AND RADIONUCLIDE PARTITION FOR NUCLEAR DEBRIS

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ADMINISTRATIVE INFORMATION

The work reported was part of a project being done for the Division of Biology and Medicine, USAEC, and sponsored under AEC Contract AT(49-7)-1963.

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ABSTRACT

This report presents a simplified, semi-theoretical model of fractionation, suitable for making interim estimates of degree of fractionation and of radionuclide partition between local, intermediate, and worldwide fallout. The principles set forth are applicable to the treatment of air-, tower-, and surface-burst debris (in the order of decreasing confidence) and to correcting fallout-prediction systems for fractionation effects. The material provides the first step necessary to illustrate theoretically the definition of contamination level proposed in Part II of this series.

SUMMARY

Problem

The effects of radionuclide fractionation severely complicate the prediction of many properties of nuclear bomb debris.

Findings

A semi-theoretical model can be used in a relatively simple manner to estimate both radionuclide fractionation and its effects on exposure dose rate. The model is recommended for illustrative purposes, rule of thumb estimates, and as a stop-gap until either better models or more extensive information becomes available.

PREFACE

This series of reports presents and discusses the effects of radionuclide fractionation in nuclear bomb debris. Part I (Ref. 3) defined fractionation as "any alteration of radionuclide composition occurring between the time of detonation and the time of radiochemical analysis which causes the debris sample to be nonrepresentative of the detonation products taken as a whole." It showed how the radionuclide compositions of fractionated samples could be correlated empirically by logarithmic relations. Part II (Ref. 1) used these relations as the basis of a technical discussion of contamination density as applied to fractionated nuclear debris. This part presents a theoretical foundation for the observed logarithmic correlations of Part I. It uses this as a simplified means of estimating fractionation as a function of particle size and the partition of product radionuclides among local, intermediate, and worldwide fallout. As an interim prediction system it is applicable, with decreasing confidence, to air-burst, tower-burst, and surface-burst debris. Part IV will extend the calculations to show how fractionation-correlation parameters can be used to estimate the exposure dose rate from nuclear debris with various degrees of fractionation. It will serve to illustrate the proposals made in Part II.

INTRODUCTION

Part II in this series[1] recommended a definition of surface density of fallout contamination which was realistically related to the spatially variable radiochemical composition in fallout patterns. In order to illustrate the principles presented in that report, one needs a means of predicting the quantities to be used in the illustration. One means of making such predictions is found in a model proposed by C. F. Miller.[2] However, Miller's model is very complex, it presents conceptual difficulties, it requires machine computation for its employment, it needs input data which is presently unknown, and it is in need of considerable, fundamental revision (cf. Ref.11). There is a definite need for a simplified, interim model for predicting the effects of radionuclide fractionation on radiation hazard from fallout.

This report describes a conceptually simple, easily usable, semi-theoretical model which is applicable to air, tower, and land-surface burst debris. It shows the relation of the model to observed correlations of fractionated debris and uses this relation to predict radionuclide partition between local, intermediate, and worldwide fallout.

In Part IV of this series, this model will form a basis for illustrating calculations of exposure-dose rate according to the newly defined surface density of contamination.

Familiarity with the preceding reports of this series[1,3] is prerequisite to understanding the material presented here.

THEORY

The model to be developed rests upon two principal assumptions. The first is that the nuclear debris consists of macroscopically homogeneous, spherical particles with a lognormal size-frequency distribution. The second is that the ultimate distribution of each mass chain among the particles is proportional to some power of the particle diameter. Additional assumptions will be required to apply the model to typical situations.

In this section we will first describe the lognormal distribution and its pertinent features. We will then apply these features to the interrelation of size, surface, and volume for a lognormal distribution of spherical particles. Finally, the relation of these features to fractionation is introduced by means of the second assumption. A relationship between pairs of mass-chain ratios is thereby obtained which is independent of particle size and in agreement with observed properties of fractionated debris.

The Lognormal Distribution

The lognormal distribution is frequently used to describe nuclear debris. Stewart[4] derived a lognormal particle size-frequency distribution from theoretical considerations, but his derivation is much less realistic when applied to a land-surface burst than it is for an air burst or tower burst. Anderson[5] has adopted a lognormal distribution of radioactivity with particle size for use in his D-Model. Miller* has proposed a lognormal distribution of mass with particle size for use in his fractionation model for land-surface bursts. It will be shown below that these proposals are in harmony. Although lognormal distributions of various types are frequently observed in samples of fallout, it remains to be established whether these distributions apply to the total amount of debris produced.

A random variable is lognormally distributed if the logarithm of the random variable is normally distributed. Thus, x is lognormally distributed with mean μ and variance σ^2 if its frequency distribution function is

$$\frac{d\Lambda(\mu,\ \sigma^2)}{d\ \ln x} = \frac{1}{\sigma\sqrt{2\pi}} \exp \left[-\frac{1}{2} \left(\frac{\ln x - \mu}{\sigma} \right)^2 \right]. \qquad (1)$$

The notation is essentially that of Aitcheson and Brown.[7] The quantity $d\,\Lambda(\mu,\ \sigma^2)/d\ \ln x$, when multiplied by $d\ \ln x$, gives the probability that a randomly chosen sample has a value of x whose logarithm lies

*C. F. Miller, Private Communication.

between $\ln x$ and $\ln x + d \ln x$. Appendix A of this report gives elementary background material on probability distributions and their relation to size-frequency distributions.

An interesting and useful property of the lognormal distribution is the following (cf. Ref. 6, Theorem 2.6). The kth-moment distribution of a Λ distribution with mean μ and variance σ^2 is also a Λ distribution but with mean $\mu + k\sigma^2$ and variance σ^2. Thus, it is easily verified that

$$x^k \frac{d\Lambda(\mu_1, \sigma^2)}{d \ln x} = \underline{x}_{12}^k \frac{d\Lambda(\mu_2, \sigma^2)}{d \ln x} \qquad (2)$$

where $\underline{x}_{12} = (\underline{x}_1 \underline{x}_2)^{1/2}$ and $\underline{x}_i = \exp \mu_i$. A useful form of Eq. 2 is obtained by writing $\mu_i = \mu + k_i\sigma^2$. Then

$$\frac{d\Lambda(\mu + k_1 \sigma^2, \sigma^2)/d \ln x}{d\Lambda(\mu + k_2 \sigma^2, \sigma^2)/d \ln x} = \left\{\frac{\underline{x}}{x} \exp\left[\left(\frac{k_1 + k_2}{2}\right)\sigma^2\right]\right\}^{k_2 - k_1}. \qquad (3)$$

<u>The Interrelation of Size, Surface, and Volume Distributions</u>

As a consequence of Eq. 2 above, if the frequency, the surface, or the volume of a collection of spherical particles is lognormally distributed with respect to the particle size, the other two quantities are also lognormally distributed with respect to the particle size. For example, let x represent the particle diameter and let its normalized frequency distribution function be $\frac{d\Lambda(\mu_n, \sigma^2)}{d \ln x}$. If N is the total number of particles and S the total surface of the particles, the

distribution of the surface with respect to x is

$$\pi x^2 N \frac{d \Lambda(\mu_n, \sigma^2)}{d \ln x} = \pi \underline{x}_{ns}^2 N \frac{d \Lambda(\mu_s, \sigma^2)}{d \ln x} \qquad (4)$$

Here $\underline{x}_s = \underline{x}_n \exp(2\sigma^2)$, so $\underline{x}_{ns} = \underline{x}_n \exp(\sigma^2)$.

Since $d \Lambda(\mu_s, \sigma^2)/d \ln x$ is normalized, the total surface is

$$S = \pi N \underline{x}_{ns}^2 . \qquad (5)$$

Similarly, the distribution of volume with x is given by

$$\frac{\pi x^3 N}{6} \frac{d \Lambda(\mu_n, \sigma^2)}{d \ln x} = \frac{\pi \underline{x}_{nv}^3 N}{6} \frac{d \Lambda(\mu_v, \sigma^2)}{d \ln x}. \qquad (6)$$

Now $\underline{x}_v = \underline{x}_n \exp(3\sigma^2)$ and the total volume is

$$V = \pi N \underline{x}_{nv}^3 \qquad (7a)$$

$$= \frac{S}{6} \underline{x}_{sv} \qquad (7b)$$

The total mass of the particles is

$$M = \rho V \qquad (8)$$

where ρ is the density. The mass is obviously distributed like the volume since the particles are homogeneous, and the mean of the mass distribution μ_m equals μ_v, and consequently $\underline{x}_m = \underline{x}_v$.

An important quantity for our purposes will be the ratio of the fraction of the total surface to the fraction of the total volume in

a given particle-size increment as a function of x. From Eq. 3 above, this ratio is

$$r_{s,v} = \frac{d \Lambda(\mu_s,\ \sigma^2)/d \ln x}{d \Lambda(\mu_v,\ \sigma^2)/d \ln x}$$

$$= \frac{\underline{x}_n}{x} \exp (5\sigma^2/2)$$

$$= \underline{x}_{sv}/x \tag{9a}$$

$$= 6\ V/Sx \tag{9b}$$

According to Eq. 9a, $r_{s,v}$ is unity when x is equal to the geometric mean of $\underline{x}_s$ and $\underline{x}_v$.

RELATION OF THE LOGNORMAL DISTRIBUTION TO FRACTIONATION

Consider now two mass chains i and j which are distributed among particles of fallout of diameter x according to x^{k_i} and x^{k_j} respectively, the fallout particles themselves having frequencies which are lognormal in x. The ratio of the fraction of mass chain i to the fraction of mass chain j in particles of size x is found from Eq. 3 to be

$$r_{i,j} = \frac{d \Lambda(\mu_n + k_i\ \sigma^2,\ \sigma^2)/d \ln x}{d \Lambda(\mu_n + k_j\ \sigma^2,\ \sigma^2)/d \ln x}$$

$$= \left\{ \frac{\underline{x}_n}{x} \quad \exp\left[\left(\frac{k_i + k_j}{2} \right) \sigma^2 \right] \right\}^{k_j - k_i} \tag{10}$$

Two ratios for two pairs of mass chains can be related by eliminating $\underline{x}_n/x$ from their respective equations:

$$r_{i,j}^{1/(k_j - k_i)} \exp\left[-\left(\frac{k_i + k_j}{2}\right)\sigma^2\right] = r_{u,v}^{1/(k_v - k_u)} \exp\left[-\left(\frac{k_u + k_v}{2}\right)\sigma^2\right]$$

or

$$\ln r_{i,j} = \frac{k_j - k_i}{k_v - k_u} \ln r_{u,v} + (k_j - k_i)(k_i + k_j - k_u - k_v)\sigma^2/2 \quad (11)$$

This equation indicates a linear relationship between the logarithms of mass-chain ratios which is independent of particle size. Linear relationships between logarithms of mass-chain ratios have in fact been observed.[3]

APPLICATION OF MODEL TO LAND SURFACE BURSTS

We will now illustrate the foregoing model by applying it to some land-surface fallout distributions currently in use at NRDL. We will compare these, first according to their physical properties, then, with the help of some additional assumptions, according to their predicted radiochemical properties. We will be particularly concerned with the variation of fractionation with particle size and with the partition of radionuclides among local, intermediate, and worldwide fallout.

The Distributions

Three lognormal distributions have been used at NRDL to describe fallout:

The N-61 distribution is used for surface bursts on Nevada soil regardless of yield. According to this distribution, half of the gross activity in the nuclear cloud is in particles with diameters of 100 μ or less and 1 % is in particles with diameters of 10^4 μ or greater.

The C-61 distribution is used for surface bursts on coral surfaces. It is also yield-invariant. According to this distribution, half of the gross activity is in particles with diameters of 100 μ or less and 1 % is in particles with diameters of 5000 μ or greater.

The distribution proposed by Miller[6] contains half of the <u>mass</u> in particles with diameters of 100 μ or less and 0.4 % of the <u>mass</u> in particles with diameters of 10^4 μ or more (1 % in particles of diameter 5600 μ or greater).

In order to illustrate the dependence of various distribution properties on the mean and variance, we will treat the N-61 and C-61 distributions as though they applied to mass (or volume, since the particles are assumed homogeneous) instead of to gross activity. Comparison of the three distributions on this basis will illustrate the sensitivity to the choice of variance. To illustrate the sensitivity to the choice of mean, we will use two distributions with the same variance as that proposed by Miller, but with mass-modal particle sizes of 50 and 200 μ. We will designate these distributions M_{50} and M_{200}, respectively, and the original Miller distribution by M_{100}.

Table 1 summarizes some important statistical and physical properties of these distributions, together with the equations involved in their

Table 1

Properties of Various Distributions, Illustrating Their Sensitivity to Mean and Variance

Quantity	C-61	N-61	M_{50}	M_{100}	M_{200}
Variance (Dimensionless)					
σ	1.682	1.980	1.736	1.736	1.736
σ^2	2.829	3.919	3.015	3.015	3.015
Modal Diameters (μ)					
$\underline{x}_v$	100	100	50	100	200
$\underline{x}_s = \underline{x}_v \exp(-\sigma^2)$	5.907	1.986	2.452	4.904	9.809
$\underline{x}_n = \underline{x}_v \exp(-3\sigma^2)$	2.04×10^{-2}	7.81×10^{-4}	5.87×10^{-3}	1.17×10^{-2}	2.35×10^{-2}
$\underline{x}_{sv} = \sqrt{\underline{x}_s \underline{x}_v}$	24.3	14.1	11.1	22.1	44.3
Means (Dimensionless)					
$\mu_v = \ln \underline{x}_v$	4.605	4.605	3.912	4.605	5.298
$\mu_s = \ln \underline{x}_s$	1.776	0.686	0.897	1.590	2.283
$\mu_n = \ln \underline{x}_n$	-3.876	-7.154	-5.137	-4.449	-3.751
Physical Properties					
$S/V = 6/\underline{x}_{sv}$ (μ^{-1})	0.247	0.426	0.541	0.271	0.135
$N/V = 6/\pi\, \underline{x}_{nv}^3$ (μ^{-3})	0.655	87.5	12.0	1.51	0.187
Fractions of Mass or Volume in Particle-Size Ranges					
0-25 μ	0.205	0.242	0.345	0.212	0.115
25-50 μ	0.135	0.121	0.155	0.133	0.097
>50 μ	0.660	0.637	0.500	0.655	0.788
Fractions of Surface in Particle-Size Ranges					
0-25 μ	0.805	0.900	0.910	0.826	0.705
25-50 μ	0.093	0.048	0.049	0.084	0.121
>50 μ	0.102	0.052	0.051	0.090	0.174
Fraction of Surface/Fraction of Volume ($r_{s,v}$) for Particle-Size Ranges					
0-25 μ	3.93	3.72	2.64	3.90	6.13
25-50 μ	0.689	0.397	0.316	0.632	1.25
>50 μ	0.155	0.082	0.102	0.137	0.221

calculation. Figure 1 shows the M_{100} distribution in differential form (frequency function) and Fig. 2 shows it in integral form (the distribution function).

We will now apply the surface-to-volume ratio to the estimation of fractionation with particle size, and will use the fractions of surface and volume in various particle-size groups to estimate the partition of individual radionuclides among local, intermediate and worldwide fallout.

Predicted Radiochemical Composition vs Particle Size

In order to apply these considerations to the estimation of radiochemical properties from a land-surface burst, two additional assumptions must now be made to obtain k values. The first is that the mass-95 chain is distributed like the volume $\left(k_{95} = 3\right)$, and the second is that the mass-89 chain is distributed like the surface $\left(k_{89} = 2\right)$. This allows $r_{89,95}$ to be equated to $r_{s,v}$ and calculated as a function of particle size from Eq. 9. The values of $r_{s,v}$ integrated over size increments are tabulated in Table 1.

Empirical correlations of radionuclide ratios have been obtained in the form[3]

$$\log_{10} r_{i,89} = a_i + b_i \log_{10} r_{95,89} \qquad (12)$$

or

$$\log_{10} r_{i,95} = a_i + (1-b_i) \log_{10} r_{89,95} \qquad (13)$$

where a_i and b_i are regression coefficients.

Comparing Eq. 13 with Eq. 11 in view of the above assumptions, $j = v = 95$, $k_j = k_v = 3$, $u = 89$, $k_u = 2$. Equation 11 becomes

$$\log r_{i,95} = (3 - k_i) \log r_{89,95} + \frac{(3 - k_i)(k_i - 2)\, \sigma^2}{2 \times 2.303} .$$

Therefore

$$b_i = k_i - 2 \tag{14}$$

$$a_i = (3 - k_i)(k_i - 2)\, \sigma^2/4.606$$

$$= b_i\,(1 - b_i)\, \sigma^2/4.606 \tag{15}$$

The evaluation of b_i by the radiochemical analysis of fallout samples or its estimate by the method described in Ref. 3, allows the radionuclide composition to be estimated as a function of particle size. If σ is known, a_i can also be calculated. For a given distribution, a_i would have a maximum value for a mass chain with b_i equal to 1/2. In such a case

$$a_{max} = \sigma^2/18.4$$

Predicted Partition of Radionuclides Among Local, Intermediate and Worldwide Fallout

The assumptions used in the preceding paragraph (viz., $k_{89} = 2$, $k_{95} = 3$) also allow estimates of the partition of various radionuclides among local, intermediate, and worldwide fallout. For this purpose we first define local fallout as consisting of all particles with diameters of 50 μ or more, intermediate fallout as particles with diameters ranging from 25 to 50 μ, and worldwide fallout as particles with diameters of 25 μ or less. The fraction of the mass-95 chain in any of these portions

is then equal to the fraction of the total particle volume in that portion, as shown in Table 1. The same is true for any other quasi-refractory mass chain (i.e., one for which $b_i \approx 1$).

The fraction of the mass-89 chain in any portion is equal to the fraction of the surface lying in that portion. Values for the three ranges cited are shown in Table 1. The same is true for any other nuclide for which $b_i = 0$.

The fraction of intermediately behaving chains can be estimated by calculating their modal volume values as in Eq. 2:

$$\underline{x}_i = \underline{x}_v \exp\left[(b_i - 1)\,\sigma^2\right] \tag{16}$$

From what has preceded it is clear that the variance for each such distribution will be the same as the variance for any geometric particle property for that distribution. Curves can then be plotted to yield the fraction in any group according to the value of b_i. This has been done for the M_{100} distribution, and the result is shown in Fig. 3. The values of b_i for some mass chains, as determined from Ref. 3, are indicated on the curve. That of Pu^{239} is estimated.

APPLICATION OF MODEL TO AIRBURSTS

As mentioned above, Stewart's treatment of nuclear debris formation is more realistic for air bursts than for land surface bursts. His calculations apply to a device composed of 1000 kg of Fe and, as can be shown from the data he uses, a yield of 10 kt. The total

yield-to-mass ratio is therefore about 10^4 (Ref. 4). For this he calculates a modal radius of about 10^{-2} μ and a particle-size frequency function

$$\frac{d\Lambda}{d \ln x} = \frac{1}{\sqrt{2\pi}} \exp\left[-\frac{1}{2} (\ln x - \mu)^2\right]. \qquad (17)$$

Stewart's variance is arbitrarily and perpetually unity. His value of $\underline{x}_n$ is

$$\underline{x}_n = \frac{v_B N_o T_o}{Kn} \left(\frac{2k}{\pi m A}\right)^{1/2} \qquad (18)$$

where

v_b = molecular volume in the liquid phase

N_o = initial number of atoms per unit volume in the fireball

T_o = absolute temperature of fireball at the time of condensation

K = $4\ kT/9\eta \approx 3 \times 10^{-9}$, T = absolute temperature, η = air viscosity

n = concentration of condensation nuclei in the fireball

k = Boltzmann's constant

m = molecular mass of iron

A = 7500° K

The particle size-frequency data available for air burst debris is not reliable below a value for x of about 5 μ. The number of particles above 5 μ represents only a small fraction of the whole, but data available are in agreement with a lognormal distribution.[7]

The assumptions of the previous section, namely, that the mass-95 chain is distributed among the particles in proportion to their volume and the mass-89 chain is distributed according to surface, are also more realistic when applied to air bursts because the debris remains in contact longer with the condensing material. The fact that fractionated samples of air burst debris correlate logarithmically[7] is also in agreement with a lognormal distribution, as explained above. Therefore, the present model can be used also in estimating fractionation for air burst debris. However, certain precautions should be indicated. Thus, Stewart's theory was developed for bursts of low yield with condensation times of the order of 10 sec. or less. No theory for longer periods has been found. However, two modifications can be made to Stewart's formula which will make it more realistic and more applicable to bursts of higher yield and lower mass.

The first modification concerns putting a lower limit on the particle size. With higher yield-to-mass ratios, smaller particles in greater abundance are to be expected. Obviously, there is a limit. In a dust-free atmosphere, it would be initially the molecular diameter. Tropospheric aerosols have a lower limit of about 8×10^{-3} μ diameter due to the coagulation of smaller particles.[8] Similar mechanisms can be expected to eventually establish a lower limit of air-burst debris particle size. In a real environment, the limit would be approximately the modal size of the dust particles. In either case, we can call this limit x_o and shift coordinates in such a manner that the distribution law does not result in smaller particles.

The second modification concerns the variation of $\underline{x}_n$ with the device's ratio of mass to total yield M/W. The value of N_o will vary as M/W. Neglecting the effect of M/W on the other parameters leads to a direct proportion between $\underline{x}_n$ and M/W. From Stewart's calculations, with M/W dimensionless,

$$\underline{x}_n \approx 10^2 \, M/W \quad \text{(microns)} .$$

Introducing this and the shift in abscissa into Eq. 17 gives

$$\frac{d\Lambda}{d \ln x} = \frac{1}{\sqrt{2\pi}} \exp\left\{ -\frac{1}{2} \left[\ln\left(\frac{x - x_o}{x_o + 10^2 \frac{M}{W}} \right)^2 \right] \right\} \qquad (19)$$

Figure 4 shows some differential graphs of this function with $x_o = 10^4 \, \mu$ for different mass-to-yield ratios. A thousand-pound device weight is assumed. Integral curves are shown in Fig. 5.

The validity of these considerations and the development of more exact equations are subjects that require further investigation and lie beyond the scope of the present report.

APPLICATION OF MODEL TO TOWER BURSTS

Stewart considers Eqs. 17 and 18 applicable to tower bursts and illustrates the application for a burst of the same yield in which 25 tons of steel tower are vaporized. For this situation he calculates $\underline{x}_n = 0.2 \, \mu$. (Our approximate equation would give $0.25 \, \mu$). Applying

the assumptions set forth above to Stewart's tower burst gives the integral curves shown in Fig. 6. As shown by the figure, the particle size of unit $r_{89,95}$ value has decreased from the value of $\underline{x}_{s,v}$ shown in Table 1 to a value of 2.5 μ. According to this treatment, therefore, a particle from a tower burst should be more depleted in Sr^{89} than a particle of the same size from a land-surface burst.

DISCUSSION

This section attempts to point out, with equal emphasis, both the defects and the merit of the present model. Although the model is a stop-gap, whose raison d'être lies more in necessity than in intrinsic worth, it will be shown that its resemblance to reality is sufficient to justify its use for illustrations, rule of thumb calculations, and estimates made in the absence of reliable information.

The principal assumptions involved in this development and application have been (1) a lognormally distributed group of homogeneous, spherical particles, (2) a distribution of mass-95 chain among the particles in proportion to the volume, and (3) a distribution of the mass-89 chain among the particles in proportion to their surface. None of these assumptions is new, and only the first is popular. The last two can be easily modified to conform to other powers of the diameter, should this appear desirable. In this section we will discuss the validity of each assumption in turn and then compare the present model with the Miller

model. Finally, the extension of the model to other burst types (water surface) will be discussed and the unifying power of the model will be explained.

Validity of the Lognormal Distribution

The case for lognormal particle-size distributions has already been discussed, but it seems appropriate to summarize it here. Stewart's prediction of lognormal distributions for nuclear debris is most validly applied to air bursts and least to land-surface bursts. The meager data available on air-burst debris is not valid below diameters of 5 μ, but data above this size can be fitted with a lognormal form and reasonable parameters obtained. Therefore, the data available support this kind of distribution but are not sufficient to substantiate it. In air bursts, the particles are extremely heterogeneous and in land-surface bursts they are more so. The properties observed must be considered as an average, and a theory that can predict the average does well.

In fallout samples from land-surface bursts, many kinds of distributions have been observed, but the lognormal activity-particle size distribution is as good an approximation to the observations as any. It must be emphasized that these samples have all been biased, not only by the sampling device itself, but also by the fallout transport process. Thus, samples collected close to ground zero were depleted in small particles and those collected at greater distances were successively depleted in large particles. No representative sample of particles from

a land-surface burst has ever been obtained, and probably none ever will be. Techniques of analyzing fallout data, now being applied at this Laboratory, may lead to more refined estimates of the initial particle-size distribution.

<u>Validity of the x^3 Distribution of the Mass-95 Chain and the x^2 Distribution of the Mass-89 Chain</u>

The assumption has been made that the mass-95 chain distributes itself throughout the fallout particles according to the cube of their diameters. In the case of air-burst debris, this seems to be a reasonable assumption. Air-burst debris has been found to be very heterogeneous with regard to color, specific activity, density, decay characteristics, and macroscopic composition, and no relationship among these properties has been determined. The activity of individual particles appears to vary approximately as the cube of the diameter for large particles, and varies progressively more like the square of the diameter as the particle size decreases.* This behavior is in qualitative agreement with the behavior expected from the present model.

With regard to land-surface bursts, two principal kinds of particles have been observed.[9,10] One kind is spherical, fractionated, of high specific activity, and the activity is distributed more or less regularly throughout the volume. The other kind is angular, less fractionated, of much lower specific activity, and the activity is concentrated on its surface. Although the latter is numerous, the former makes the larger contribution to the radiological field and contains

*Leon Leventhal, private communication

the larger fraction of the mass-95 chain. The former kind appears to be comprised of particles that were completely melted and perhaps partially vaporized. The larger of these presumably fell away from the nuclear cloud while the mass-89 chain was predominantly in the uncondensible Kr form. The large, irregular particles are apparently formed by the indiscriminate condensation of vapors on cold particle surfaces or by the scavenging of small, condensed particles. Fractionation was observed to cause the quantity of the mass-89 chain carried by a particle to be independent of the particle type, by counteracting the increased activity of regular particles with Sr^{89} depletion.

A further piece of information on the validity of the distributions chosen for the mass-95 and mass-89 chains has recently appeared in a report by J. H. Norman and W. E. Bell.[11] On the basis of their studies of Cs vaporization from, and condensation on, silicate melts of varying compositions, these authors conclude: "Those elements that exhibit relatively low vapor pressures will condense onto fallout particles at high temperatures where rates of diffusion are relatively high.... It is expected that these elements will readily diffuse into fallout particles and will become fairly uniformly distributed. On the other hand, elements that exhibit relatively high vapor pressures will condense onto fallout particles at low temperatures, where rates of diffusion are low. It is expected that these elements will be retained on or near the surface of the particle."

It therefore appears that, in the absence of agglomeration, there are reasonable distributions to expect, but that further study is required to take agglomeration effects into account.

Comparison with the Miller Model

The Miller model and the present one have certain similarities. Both adopt a lognormal mass vs. particle-size distribution and, if the particles are considered to be completely melted, both will predict that refractorily behaving mass-chains are distributed among the particles in proportion to the cube of the diameter.

The Miller model has the advantages of accounting for particles which leave the cloud before the mass-89 chain has condensed and of being able to handle partially melted particles. This is good with regard to the prediction of refractorily behaving chains, but bad with regard to the prediction of volatilely behaving chains. Thus, the closest that one comes to accounting for the mass-89 chain contributed by unfractionated, irregular particles is to approximate them with partially melted spherical particles depleted in Sr^{89}.

The present model tends to correct this defect. Whether the extent of correction is so unrealistically large that it is better omitted remains to be seen.

Unitive Power of the Present Model

The applicability of the present model to air, tower, and land-surface bursts is a point in its favor. It has been pointed out[12] that

the condensation processes occurring when a nuclear device is detonated on the surface of a body of fresh water can be approximated by those of an air burst occurring in the presence of vaporized water. The present model should provide orientation for considering this type of situation. In the case of an ocean-surface burst, the situation is similar but complicated by the presence of vaporized sea salts.

All these situations will receive consideration in due course.

<u>Application to Models for Fallout Transport</u>

There exist many models for calculating the transport of radioactive fallout from the nuclear cloud to the ground. Many of these consider the debris to be divided into particles of different sizes. By means of the model described here, the degree of fractionation can be estimated for any particle-size range. The next report of this series will illustrate the estimation of exposure dose-rate as a function of time and degree of fractionation. With calculations of this type, the contribution to the dose-rate from each particle class can be estimated and fractionation accounted for.

<u>Resemblance of Model Predictions to Observations of Nuclear Debris</u>

Without involving classified material, the resemblance of model prediction to fallout observations may be summarized as follows:

(1) The model is in agreement with observed correlations of radionuclide composition for fractionated debris in both air and surface bursts.

(2) The model gives the observed trend in radionuclide composition

with particle size (greater enrichment in volatile-behaving chains with decreasing particle diameter) in both air and surface bursts.

(3) In a 2-kt, near-surface burst, the particle diameter for which $r_{89,95} = 1$ was observed to be 27 ± 17 μ, in good agreement with the values of $\underline{x}_{s,v}$ shown in Table 1. However, particles greater than several hundred microns were found to be less depleted in volatilely behaving chains than the model predicts. This departure may be due to the incorporation of smaller, more volatile-rich particles on these larger particles by impaction. Evidence of impaction also appeared in microscopic and autoradiographic examinations of this debris.

(4) The model predicts reasonable partition between local and worldwide fallout.

(5) The model offers a significant improvement over the use of unfractionated radionuclide compositions.

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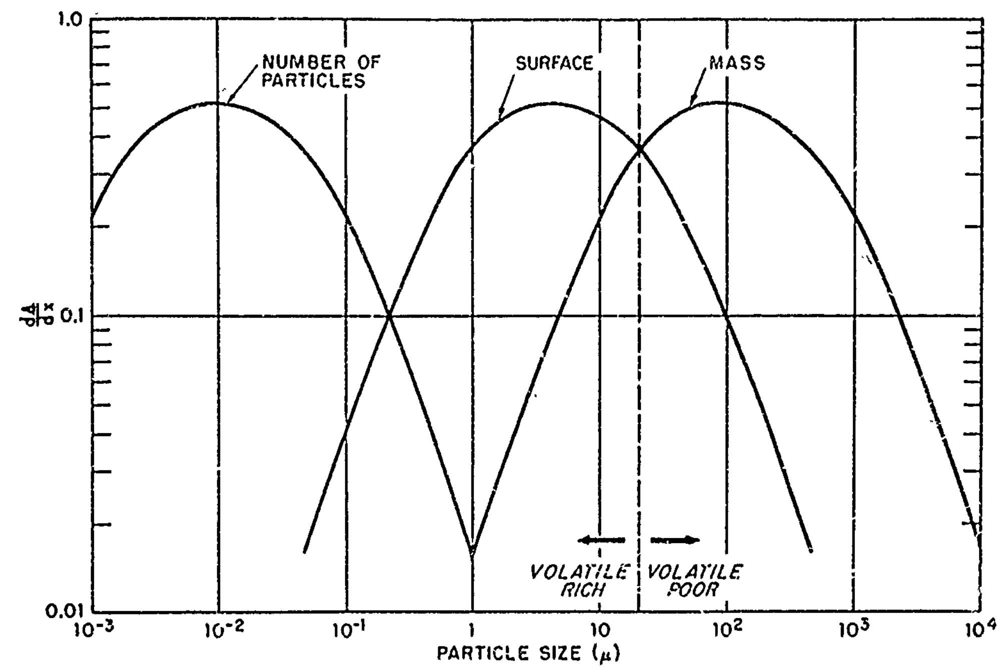

Fig. 1 Frequency Function (Differential) Curves of Mass, Surface and Number of Particles as a Function of Particle Diameter for the M_{100} Distribution. The figure shows the shift in modal value with distribution moment and the occurrence of x_{sv} where the surface and volume curves intersect.

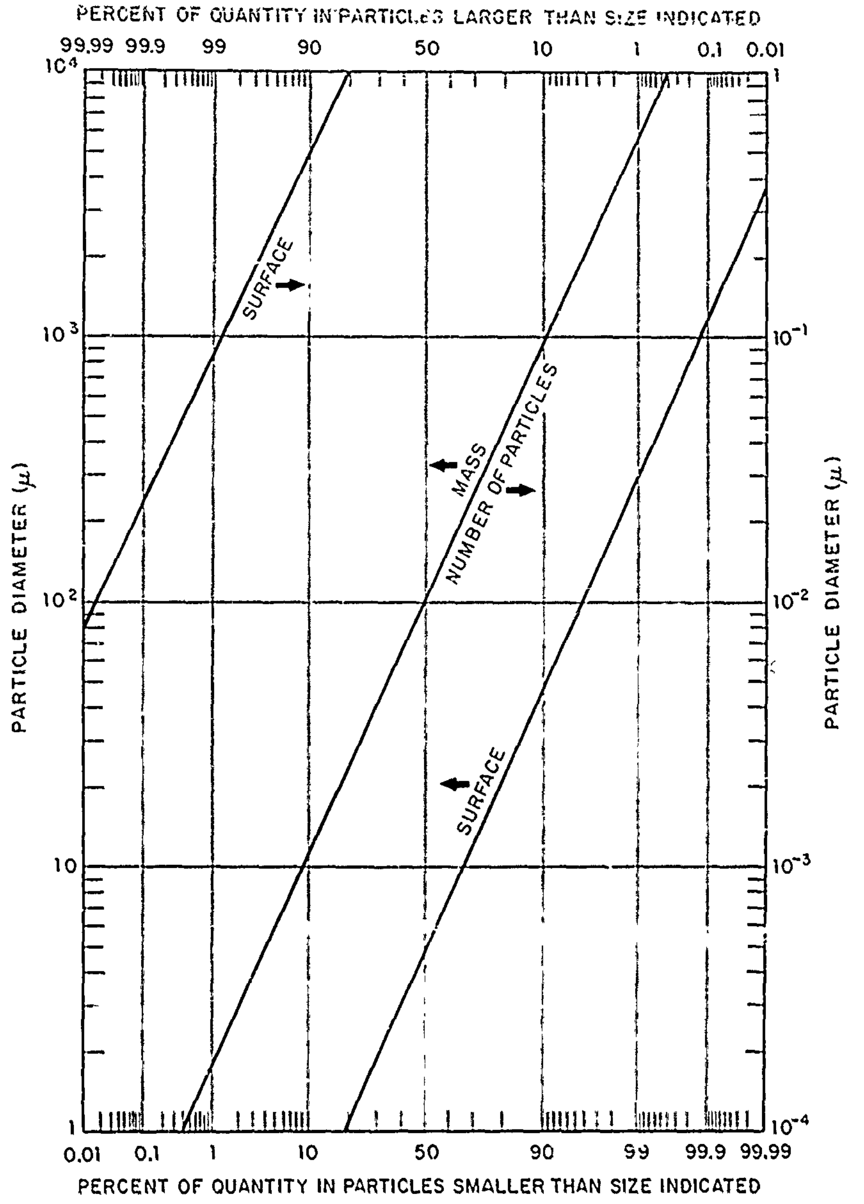

Fig. 2 Distribution Function (Integral) Curves of Mass, Surface and Number of Particles as a Function of Particle Diameter for the M_{100} Distribution

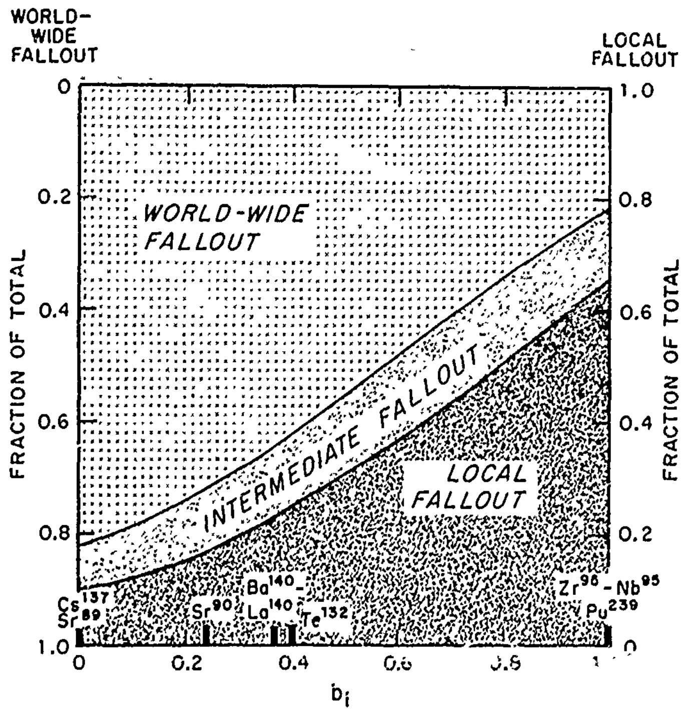

Fig. 3 Partition of Mass Chains Among Local, Intermediate and Worldwide Fallout as a Function of b_i for the M_{100} Distribution

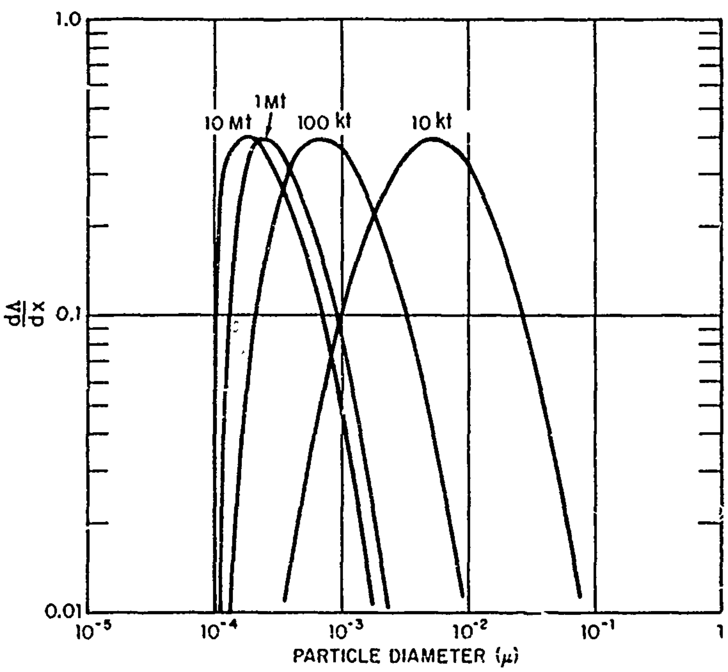

Fig. 4 Particle Size-Frequency Function (Differential) Curves for Air Bursts of Various Mass-to-Yield Ratios for $x_o = 10^{-4}\ \mu$

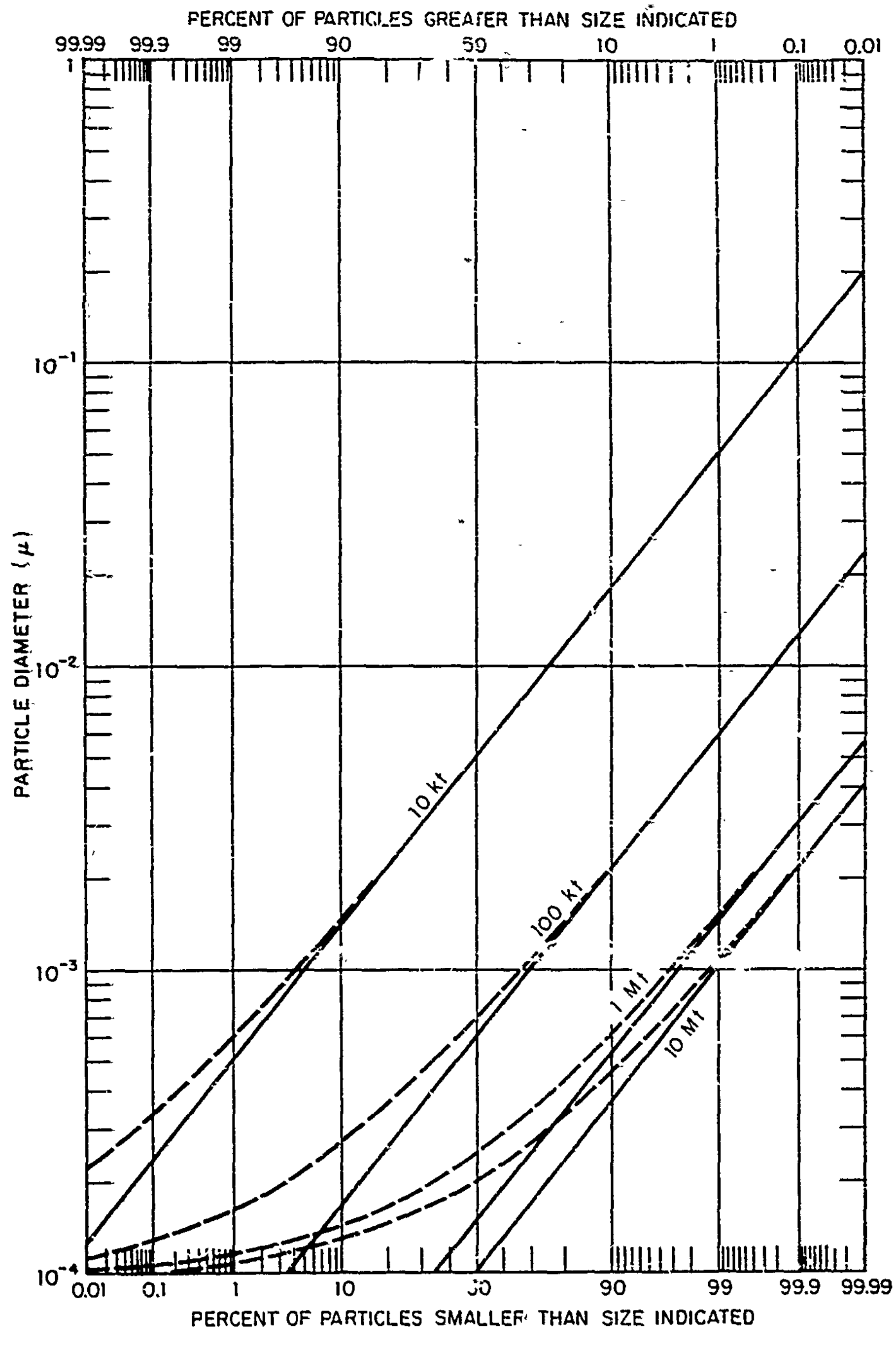

Fig. 5 Particle Size Distribution Function (Integral) Curves for Air Bursts of Various Mass-to-Yield Ratios*

*Solid lines are plotted for $x - x_0$. Dashed lines are plotted for x, assuming $x_0 = 10^{-4}$ micron

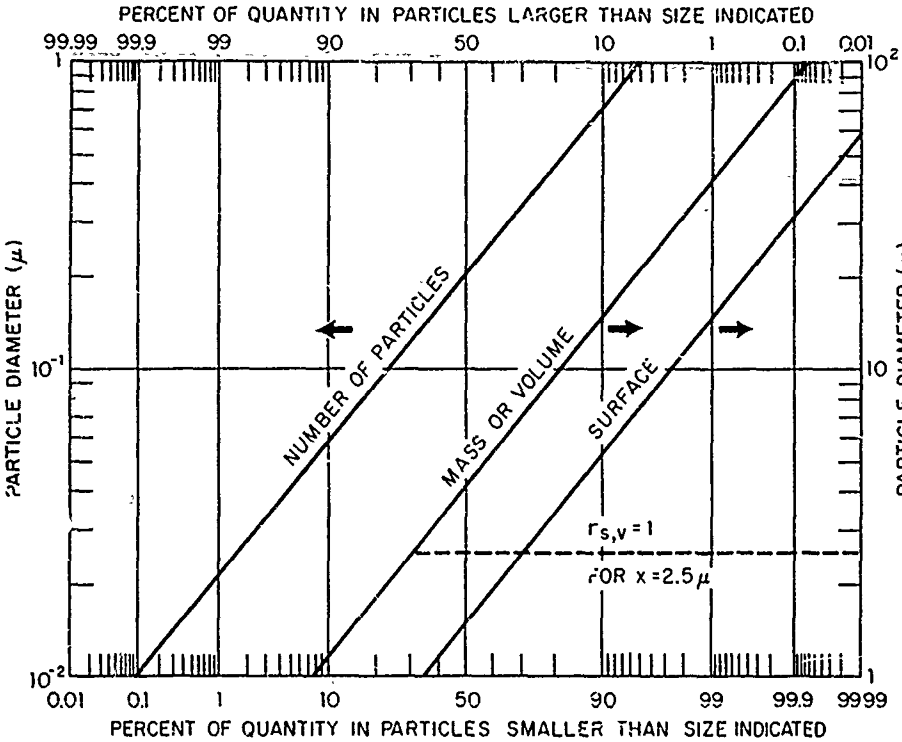

Fig. 6 Particle Size Distribution Function (Integral) Curves for a 10-kt Tower Burst Vaporizing 25 t of Steel

APPENDIX A

RELATION OF SIZE-FREQUENCY DISTRIBUTIONS TO PROBABILITY DISTRIBUTIONS

If we let $N\{a,b\}$ be the number of particles in a given sample whose diameters x satisfy the inequality $a < x \leq b$, then $N\{0,\infty\}$ is the total number of particles in the sample and $N\{a,b\}/N\{0,\infty\} = P\{a,b\}$, the probability that a randomly chosen particle has a diameter x such that $a < x \leq b$. The size frequency function $p(x)$ is related to this probability by

$$\left.\begin{aligned} P\{a,b\} &= \int_a^b p(x)dx \\ \frac{dP}{dx} &= p(x) \end{aligned}\right\} \qquad \text{(A1)}$$

If it is desired to approximate the size-frequency distribution with a normal distribution, this is easily accomplished with the standard normal distribution function

$$y = \frac{1}{\sqrt{2\pi}} \exp(-t^2/2) \qquad \text{(A2)}$$

by making the equality

$$p(x)dx = y\,dt = y\,\frac{dt}{dx}\,dx$$

from which

$$p(x) = y\,\frac{dt}{dx} \qquad \text{(A3)}$$

Thus, for a normal distribution:

$$t = \frac{x - \mu}{\sigma},$$

$$\frac{dt}{dx} = \frac{1}{\sigma},$$

$$y = \frac{1}{\sqrt{2\pi}} \exp\left[-\frac{1}{2}\left(\frac{x - \mu}{\sigma}\right)^2\right],$$

$$p(x) = \frac{1}{\sigma\sqrt{2\pi}} \exp\left[-\frac{1}{2}\left(\frac{x - \mu}{\sigma}\right)^2\right]. \qquad (A4)$$

For a lognormal distribution

$$t = \frac{\ln x - \mu}{\sigma},$$

$$\frac{dt}{dx} = \frac{1}{x\sigma},$$

$$y = \frac{1}{\sqrt{2\pi}} \exp\left[-\frac{1}{2}\left(\frac{\ln x - \mu}{\sigma}\right)^2\right],$$

$$p(x) = \frac{1}{x\sigma\sqrt{2\pi}} \exp\left[-\frac{1}{2}\left(\frac{\ln x - \mu}{\sigma}\right)\right]^2.$$

$$= \frac{d\Lambda(\mu, \sigma^2)}{dx} \qquad (A5)$$

Note this is not the $d\Lambda/d \ln x$ as in Equation (1).

The lognormal distribution has been treated at length by several authors (A1-A3) besides Aitcheson and Brown, but it will be helpful to summarize some of its essential properties here. A random variable

is distributed lognormally if the logarithm of the variable is distributed normally. Such a variable can only have positive values. The value of the variable x for which the frequency is a maximum is the modal value $\underline{x}$ and its natural logarithm

$$\ln \underline{x} = \mu$$

is the mean value of the ln x. We will call the values of x for which $t = \pm 1$ by x_{+1} and x_{-1}. At these values, y falls to $\exp(-\frac{1}{2})$ of its maximum value $\frac{1}{\sqrt{2\pi}}$, $-d \ln y/dt$ is unity, and $d^2y/dt^2 = 0$. The standard deviation σ is given by $\ln x_{+1} - \ln \underline{x} = \ln \underline{x} - \ln x_{-1}$, the difference between the values of ln x at the axis of symmetry and the points of inflection. Thus t represents the departure, in σ units, of ln x from its mean value.

Finally,

$$p(x)\,dx = \frac{1}{\sqrt{2\pi}} \exp\left[-\frac{1}{2}\left(\frac{\ln x-\mu}{\sigma}\right)^2\right]\left(\frac{d \ln x}{\sigma}\right)$$

$$= P(x, x + dx)$$

$$= \frac{d \Lambda(\mu, \sigma^2)}{d \ln x}$$

is also the probability that ln x lies between ln x and ln x + d ln x.

Figure 7 illustrates several presentations of the same distribution and their relation to one another. Figure 7A is a logarithmic-probability presentation for Y when $\underline{x} = 1$ and $x_{+1} = 1/x_{-1} = 2$. The linear-probability presentation of the normal distribution obtained by letting x' = ln x

is shown in Figure 7B. Here, $\underline{x} = 0$, $x_{+1} = -x_{-1} = \sigma = 0.693$. This is converted to the more familiar frequency function in Figure 7C and to the less familiar Gaussian plot (A4). These in turn are converted back to the logarithmic form in Figure 7D.

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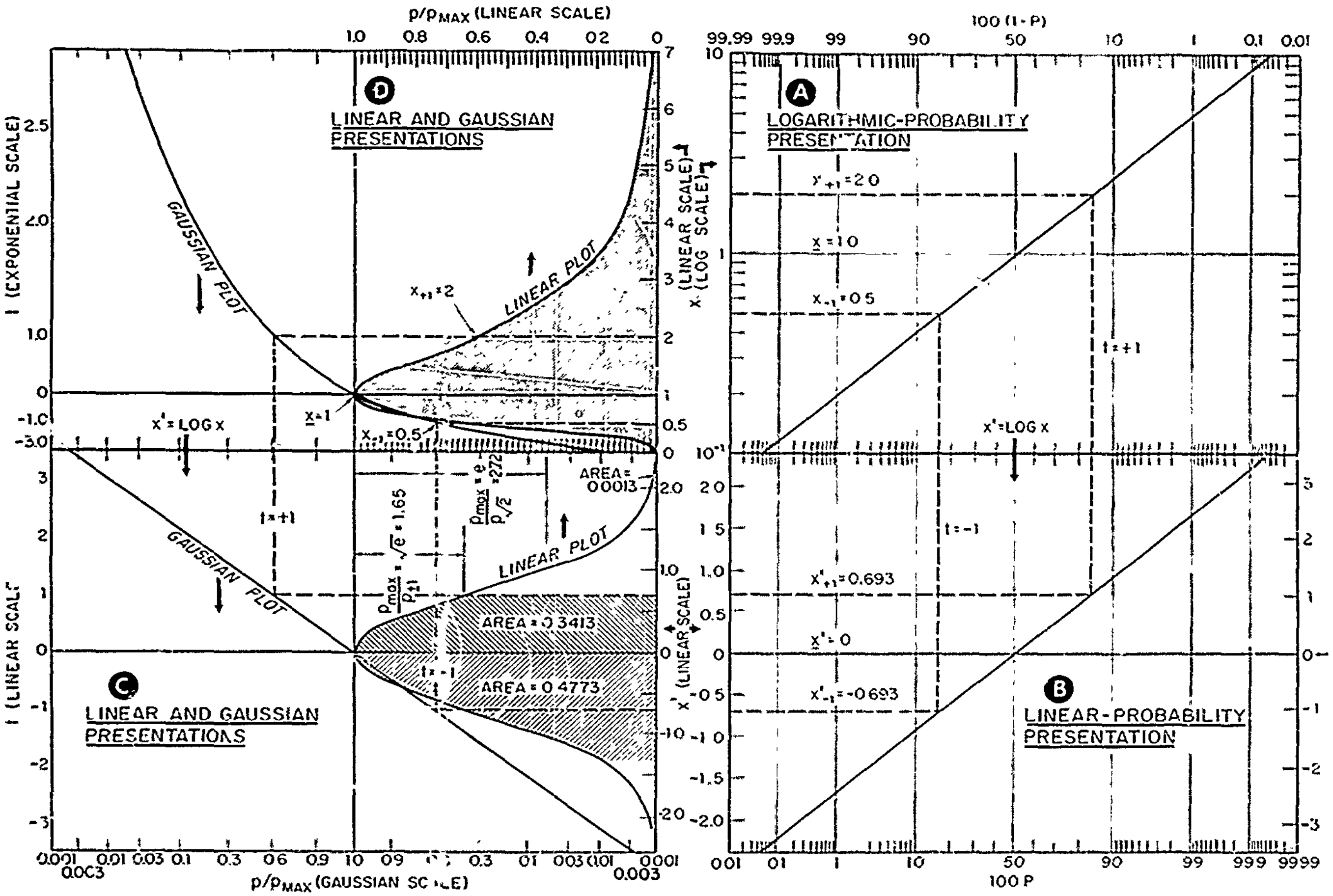

Fig. 7 Interrelation of Various Presentations of the Lognormal Distribution

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FRACTIONATION III. ESTIMATION OF DEGREE OF FRACTIONATION AND RADIONUCLIDE PARTITION FOR NUCLEAR DEBRIS by E.C. Freiling
12 September 1963 46 p. table illus. 16 refs.
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This report presents a simplified, semi-theoretical model of fractionation, suitable for making <u>interim</u> estimates of degree of fractionation and of radionuclide partition between local, intermediate, and worldwide fallout.

(over)

1. Fractionation.
2. Dose rate.
3. Radiological contamination.
4. Radioactive fallout.
5. Mathematical prediction.
6. Particles.
7. Particle size.
I. Freiling, E.C.
II. Title.

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The principles set forth are applicable to the treatment of air-, tower-, and surface-burst debris (in the order of decreasing confidence) and to correcting fallout-prediction systems for fractionation effects. The material provides the first step necessary to illustrate theoretically the definition of contamination level proposed in Part II of this series.

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